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Modeling the homogenization kinetics of as-cast U-10wt% Mo alloys

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ABSTRACT

Low-enriched U-22at% Mo (U–10Mo) alloy has been considered as an alternative material to replace the highly enriched fuels in research reactors. For the U–10Mo to work effectively and replace the existing fuel material, a thorough understanding of the microstructure development from as-cast to the final formed structure is required. The as-cast microstructure typically resembles an inhomogeneous microstructure with regions containing molybdenum-rich and -lean regions, which may affect the processing and possibly the in-reactor performance. This as-cast structure must be homogenized by thermal treatment to produce a uniform Mo distribution. The development of a modeling capability will improve the understanding of the effect of initial microstructures on the Mo homogenization kinetics. In the current work, we investigated the effect of as-cast microstructure on the homogenization kinetics. The kinetics of the homogenization was modeled based on a rigorous algorithm that relates the line scan data of Mo concentration to the gray scale in energy dispersive spectroscopy images, which was used to generate a reconstructed Mo concentration map. The map was then used as realistic microstructure input for physics-based homogenization models, where the entire homogenization kinetics can be simulated and validated against the available experiment data at different homogenization times and temperatures.

1. Introduction

Enriched uranium (>90% ^{235}U) has been widely used in many nuclear applications where high fissile material density is needed. The proliferation concern associated with the use of highly enriched uranium (HEU: enrichment >20% ^{235}U) motivates the U.S. Department of Energy's (DOE's) United States High Performance Research Reactor (USHPRR) Conversion Program to seek low-enriched uranium (LEU) alternatives. The conversion of HEU research reactors to the use of LEU requires an increase in the total density of uranium atoms in the fuel. The previous scoping studies identified uranium–molybdenum (U–Mo) alloys with Mo concentration 7–10 wt% as promising candidate materials for LEU fuel because of their favorable attributes under irradiation and high intrinsic uranium density [1,2]. Gamma-phase U with Mo in solid solution possesses acceptable irradiation stability, good mechanical properties and corrosion resistance, and can be formed over a wide range of Mo concentration.

Two forms of U–Mo alloy fuel plates have been investigated and tested under the USHPRR Conversion Program: a dispersion fuel with fuel particles dispersed in an aluminum matrix, and a monolithic fuel in aluminum alloy cladding [2,3]. The fuel plate assembly consists of a central layer (fuel meat with U–Mo alloy) sandwiched between cladding sheets, and is manufactured by various hot and cold rolling processes.

Complex materials processing techniques including casting, thermal annealing, hot and cold rolling, coating, and hot isostatic pressing (HIP) are used to fabricate U–Mo metallic fuel plates. Based on the earlier work [1–7] on the thermomechanical processing of U–10Mo it was determined that different combinations and sequences of material processes may result in a variety of microstructures with different Mo segregation, grain morphology, phase volume fractions, carbide inclusions, and so on. In order to fabricate U–10Mo monolithic plates consistently, at a lower cost and with high yield, a thorough understanding of the microstructure evolution kinetics from as-cast to the final formed

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structure after various material processes is required. The U–10Mo as-cast microstructure is an inhomogeneous dendritic structure with molybdenum-rich and -lean regions [4–7]. It has been observed in U–10Mo that, depending on the casting process adopted [5], microstructures with different grain size, molybdenum inhomogeneity, secondary dendrite arm spacing and carbide distribution were observed. Mo segregation during casting may affect γ phase stability and the formation of α phase as well as the phase transition from γ to α and γ' during thermal annealing. A homogenization process is needed to alleviate Mo segregation and produce desired microstructures with uniformly distributed Mo. Homogenization of U–10Mo is performed in the γ-phase field (above 560 °C) for several hours [7–9]. The kinetics of the homogenization is dependent on the grain size and the other aforementioned parameters, and experimentally determining the time and temperature to homogenize is time-consuming and labor-intensive. Recent work on homogenization of U–10Mo was performed by Leenaers [7] and Joshi et al. [4–6]. The homogeneity or inhomogeneity was observed using the x-ray techniques in scanning electron microscope (SEM). The as-cast samples in these cases were annealed for several hours and the microstructure was observed for inhomogeneity. In order to overcome this heuristic approach, a homogenization model is necessary to predict the kinetics for varied grain sizes. Computational modeling combined with experimental validation will improve understanding of microstructural evolution kinetics and make it possible to improve the alloy design with optimized process parameters to achieve desired microstructures and material properties.

Prior work involving the modeling of the homogenization kinetics in the as-cast U–10Mo does not exist; however preliminary work involving U–Nb alloy work was performed by Snyder in 1978 [10,11] and more recently by Lopez et al. on U-7.5Nb-2.5Zr [12] alloys. These models used scanning electron microscope (SEM) energy dispersive spectroscopy (EDS) line scans and microhardness measurements to determine the degree of inhomogeneity. A simple analytical model [13] was used by Snyder [6] to determine the time required to homogenize the entire casting. Despite the ease of using this model, that work does not take into account the variability associated with the entire microstructure or is localized to the regions where the line scans are taken and the step size of the scans. A better way to include the entire microstructure is to perform microprobe analysis on the entire image (wavelength dispersive spectroscopy [WDS] maps). This technique, though reliable, is tedious and time-consuming, and techniques that better correlate the microstructure with chemistry are required. Other techniques based on phase field, calculation of phase diagrams, etc. [14–18], to simulate the homogenization kinetics for different alloys are present in the literature [19–21]. These techniques use special tools or software that usually require a specialist and resources that are uncommon in the production environment.

The objective of this paper is to present a homogenization model of the U–10Mo alloy. To model the homogenization of U–10Mo, transport processes taking place in the metal, along the grain boundary, and from bulk alloy to grain boundary must be considered in detail. Such transport processes should have important roles in defining the overall kinetic rate of homogenization. The paper is organized as follows: an experimental study of homogenization in U–10Mo is presented first in Section II, followed by description of a modeling study of homogenization kinetics in Section III. Section IV applies the proposed model to homogenization of U–10Mo with results and discussion. Connections to experiments are presented where possible, and conclusions are drawn in Section V.

2. Homogenization studies for Idaho National Laboratory (INL) pin casting samples

2.1. Experimental procedure

For the current work, U–10Mo samples were machined from 38.1 mm-long by 6.35 mm-diameter pins that had been arc melted and cast into copper molds at INL, Idaho Falls, USA. The feedstock used in the arc melting was in the form of pieces sectioned from a coupon that had been machined from a vacuum-induction-melted and cast plate of U–10Mo. The plate was melted at 1330 °C, cast into a graphite mold and homogenized in-mold at 1100 °C for 4 h. All materials processing, other than that performed at INL, was conducted at the Y-12 National Security Complex, Oak Ridge, USA. The bulk chemical composition of the arc-cast samples was not measured; however, the composition of the coupon was determined to be U 88.7 wt% (99.8 wt% ^{238}U), Mo 10.1 wt%, C 0.08 wt%, and Si 0.04 wt%, with Al, Co, W and Cu accounting for less than 1.5 wt%. The homogenization heat treatments were conducted in a high-temperature vacuum furnace (MTI Model VBF-1200X) operated under inert atmosphere using high-purity bottled argon. The atmospheric flow rate was maintained at approximately 11.7×10^{-6} m^3/s. The samples were wrapped in Zr foil and placed in an alumina crucible (with lid). The furnace was evacuated and backfilled with argon three times; then continuous argon flow was used to establish a slight positive pressure. The heating cycle used a 10 °C/min ramp rate to 350 °C, a 15 min hold, and another ramp at the same rate to the homogenization temperature for the required amount of time, i.e., 4–48 h. This was followed by furnace cooling under flowing argon back to room temperature. The cooling rate was approximately 25 °C/min.

For the microstructural characterization, samples were cold-mounted in an epoxy resin and then sectioned at approximately half the length. The mounted samples were successively polished down to 600 grit with SiC polishing papers and were further polished using 6 μm and 1 μm diamond slurries, followed by a final colloidal silica polish. Upon polishing, the samples were gold coated for SEM analysis. The detailed technique used to prepare the samples for characterization can be found elsewhere [22]. Microstructural characterization was performed using an optical microscope as well as using a JEOL JSM-7600F SEM equipped with an Oxford Instruments X-Max 80 EDS detector. The EDS analysis was performed using the INCA Microanalysis Suite software, version 4.15. For the line scan analysis, the heat-treated samples were imaged at 500 × in backscattered electron (BSE-SEM) mode to highlight the atomic number contrast (Z contrast). Three EDS line scans of 225 μm length were performed at random regions. The line scans were used to count the molybdenum content for 30 s at an interval of 0.9 μm. Due to the presence of carbides and oxygen in the system, the EDS line scans also involved detecting these elements. Due to rapid oxidation of the surface of the U–10Mo sample upon polishing and the presence of adventitious carbon and the small interaction volume of the electron beam; the composition of the molybdenum is skewed. However this being a systematic error will not affect the eventual data.

2.2. Results (800 °C and 1000 °C): BSE-SEM images and line scan data

In order to determine the time and temperature required to transform the cast structure to the homogenized structure, a series of heat treatments were conducted. The samples were homogenized at 800 °C for 4, 8, 16, 24 and 48 h [23]. Fig. 1a, b, c are the typical as-cast, 800 °C for 4 h, and 800 °C for 24 h, respectively, of U–10Mo alloy in the BSE-SEM mode. The chemical homogeneity